Charge collective modes in an incommensurately modulated cuprate

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We report the first measurement of *collective charge modes* of insulating $Sr_{14}Cu_{24}O_{41}$ using *inelastic* resonant x-ray scattering over the complete Brillouin zone. Our results show that the intense excitation modes at the charge gap edge predominantly originate from the ladder-containing planar substructures. The observed ladder modes (E vs. Q) are found to be dispersive for momentum transfers along the "legs" ($\vec{Q} \parallel \hat{c}$) but nearly localized along the "rungs" ($\vec{Q} \parallel \hat{a}$). Dispersion and peakwidth characteristics are similar to the charge spectrum of 1D Mott insulators, and we show that our results can be understood in the strong coupling limit ($U \gg t_{ladder} > t_{chain}$). The observed behavior is in marked contrast to the charge spectrum seen in most two dimensional cuprates. Quite generally, our results also show that momentum-tunability of inelastic scattering can be used to resolve mode contributions in multi-component incommensurate systems.

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The evolution of charge dynamics with dimensional cross-over is one of the most fundamental themes in strongly correlated electron systems. For example, in 1D systems electrons exhibit charge-spin separation [1] and in 2D superconductivity appears upon doping [2]. The electron behavior in materials with somewhat *intermediate dimensionality* is poorly understood but of great current interest [3, 4, 5, 6, 7]. The doped cuprate compound $Sr_{14-x}Ca_xCu_{24}O_{41}$ series exhibits unusual quantum magnetism, electron-phonon coupling, charge-order and superconductivity under high pressure (T_c up to $\sim$ 12K at $P > 3$ GPa) and shares many unconventional properties that are also observed in the 2D cuprates [4, 5, 6, 7]. Unlike 1D (Sr_2CuO_3 or $SrCuO_2$) or 2D (LSCO or NCCO) cuprates, the structurally incommensurate cuprate system $Sr_{14}Cu_{24}O_{41}$ (SCO) consists of two Cu-O structural units as stacked planes of chains and two-leg ladders. Photoemission spectroscopy, which gives a measure of the momentum-resolved band structure, is limited due to charging, cleavage and surface issues on this entire class of compounds[8]. Therefore, unlike most other copper oxides where photoemission spectroscopy works quite well [9], very little is known about the *momentum-resolved electronic structure and excitations* of this unusual cuprate class.

Rapid recent developments show that momentum-resolved inelastic x-ray scattering is highly sensitive in distinguishing between propagating charge excitations in chain-like (1D) [10, 11] and planar (2D) [12] networks. It is also capable of revealing the nature of the electronic correlations via the measurement of detailed $\boldsymbol{Q}$-dependence of the excitations along different crystallographic directions [10, 11, 12, 13]. Such measurements provide the essential components for identifying an effective theory of electronic behavior of correlated insulators [2, 3, 10, 11, 12, 13, 14]. The inelastic x-ray charge spectrum of 1D [10, 11] cuprates exhibits radically different behavior from that of the 2D [12] cuprates. In 1D, a strongly dispersive holon resonance [10] with half-periodic spinon-like onset is observed [11] whereas in 2D exciton-like modes with Zhang-Rice character dominate [12]. Therefore, it is of interest to study the modes of a system with intermediate dimensionality of its own right to understand the nature of the excitations. In this Letter, we present an inelastic resonant x-ray scattering study of $Sr_{14}Cu_{24}O_{41}$ to elucidate the nature of the interplay between the reduced dimensionality, lattice substructures and electronic correlations by exploring the Q-space (for the first time). A systematic analysis of our data suggests that the electron-hole pair modes in $Sr_{14}Cu_{24}O_{41}$, in the vicinity of the gap edge, predominantly originate from the quasi-two dimensional ladder planes, and in some respects resemble collective charge motion observed in 1D cuprates rather than that found in the 2D systems where superconductivity is observed.

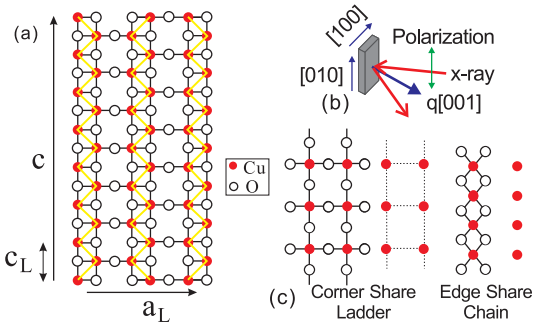

FIG. 1: **Lattice substructures and scattering geometries**: (a) Ladder unit cell in $Sr_{14}Cu_{24}O_{41}$. The bonding between two adjacent ladders (yellow lines) is very weak. (b) The scattering geometry is configured with E-field aligned in the [010] (b-axis) direction. (c) Two distinct Cu-O substructural units are shown. These fundamental units are not commensurate with each other ($7c_L \sim 10c_{ch}$).

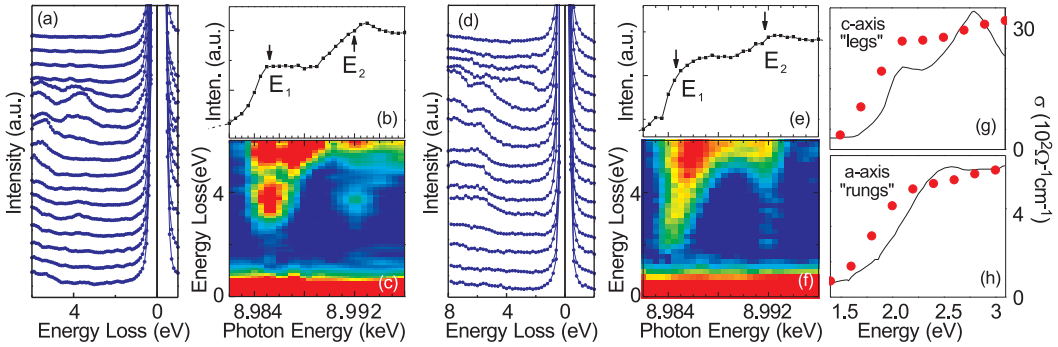

FIG. 2: **Inelastic Resonance profiles**: (a) Energy dispersion curves for incident energy dependence of $SrCuO_2$ at $Q = \pi$, with hν from 8.980 keV (top) to 8.996 keV (bottom). (b) Observed resonance energies are labeled on the Cu k-edge fluorescence spectrum. (c) Two resonance energies that couple to valence band charge excitations are visible in the incident energy dependence image plot. Energy dispersion curves for $Sr_{14}Cu_{24}O_{41}$ at $Q = 2\pi/c_L$ with hν from 8.982 keV to 8.996 keV (d) also show two resonances, labeled on a plot of fluorescence (e). RIXS data for $Sr_{14}Cu_{24}O_{41}$ at 8.984keV incident energy and $Q = 2\pi$ along the $\hat{a}$ (g) and $\hat{c}$-axes (h) with polarization along the $\hat{b}$ axis, are compared with optical conductivity data [6]. The on-set of excitation DOS is in good agreement with x-ray response for Q $\sim 2\pi$ (BZ center).

The compound $Sr_{14}Cu_{24}O_{41}$ contains two copper oxide planar sublattices, one composed of Cu_2O_3 two leg ladders (Fig-1(a)) and one forming edge-sharing CuO_2 chains. These lattices are incommensurate in the axial (c_L) direction, and roughly seven ladder unit cells correspond to ten chain units ($7c_L \sim 10c_{ch}$). The system is also inherently hole doped, however it has been suggested that most holes reside localized on the chain planes [6]. The samples were grown using an optical floating zone method and the experiments were performed at ambient temperatures and pressure using the high flux undulator beamline 9-ID at the Advanced Photon Source. Inelastic scattering was measured by varying Q along the $\hat{a}$ and $\hat{c}$ axes of single crystalline $Sr_{14}Cu_{24}O_{41}$. The chain compound $SrCuO_2$ was studied for comparison with the excitation spectra of $Sr_{14}Cu_{24}O_{41}$, since no data were previously reported for the incident energy dependence of the mode spectrum of $SrCuO_2$ [10]. Incident polarization for all spectra was maintained perpendicular to the Cu-O plaquette. The scattered beam was reflected from a diced Ge(733)-based analyzer for energy analysis and focused onto a solid-state detector that was thermoelectrically cooled to achieve a low level of background noise. Under these configurations, the experimental apparatus achieved a resolution of 120 meV, sufficient to resolve significant features across the insulating gap.

Coupling of the x-ray scattering process to specific excitations relies on resonance with intermediate charge excitations, and is dependent on the incident photon energy. Therefore, we first thoroughly investigate the detailed incident energy behavior of the excitation modes, and the summary of results is shown in Fig-2. The lower energy spectral weight corresponding to the optical gap (charge-transfer or effective Mott gap) is enhanced while incident x-ray energies are set near E_1= 8.9845keV or E_2=8.992keV. For low dimensional systems, *particularly in 1D*, it is generally found that if incident x-ray energy is tuned near the absorption peak (E_1 at the c-geometry, Fig-1), electron-hole excitations near the insulating gap-edge (similar to an optical-edge, Fig-2(g,h)) are excited and the broad momentum dispersion (E vs. Q) of the excitations reflects the excitation modes expected in the charge dynamic response, although the lineshape and cross-section effects vary [10, 11, 12, 13] in details. The incident energy dependent inelastic profile of the ladder (Fig-2) is qualitatively similar to that of the chain $SrCuO_2$ which has been shown to exhibit [11] a broad representation of E vs. Q expected in the intrinsic charge dynamic response based on DMRG calculations [11]. This was also shown to be the case in the numerical ED studies of Hubbard model in 1D by Tustsui et.al.[13].

Momentum dependence along the $\hat{c}$-direction is shown in Fig-3(a-b) as measured near the E_1 resonance of $Sr_{14}Cu_{24}O_{41}$. An upward slope in the direction of greater energy loss is seen in all curves. These higher energy spectral features arising out of various interband transitions, appear to be almost dispersionless, and have been subtracted when fitting lower energy spectral weight. A broad, flat topped feature is seen between $2eV$ and $4.5eV$ in all curves, and can be fitted well with two 1.2eV width Lorentzians separated by $\sim 1eV$, as shown at the bottom of the figure. The low energy peaks are slightly more distinct at 8.984keV than 8.9845, however a feature at 5.3eV energy loss is only clearly visible at 8.9845keV. A dip between these features is visible in the BZ center and at the zone edge. Tracing the peaks (red lines in Fig-3(a)) yields a dispersion of $\sim 1eV$ which is larger than what is typically observed in 2D prototype cuprates[12], but similar to the dispersion of $SrCuO_2$. Fig-3(c) shows momentum transfer along the $\hat{a}$-direction

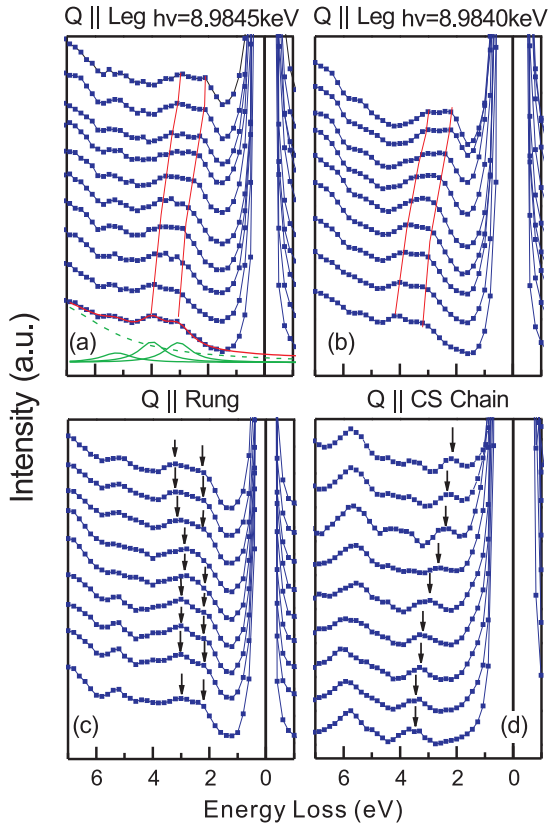

FIG. 3: **Q-dependence of charge modes**: The dispersion of low energy features studied with incident energies of 8.884keV (a) and 8.9845keV (b) is traced in red over energy dispersion curves for momentum transfer in the ladder direction, from $Q = 2\pi/c_L$ (top) to π/c_L (bottom). (c) Low energy features (black arrows) have no measurable dispersion in the $\hat{a}$ direction, shown from $Q = 6\pi$ (top) to 5π with incident energy 8.9845keV. Intensity scale is smaller than (b) by a factor of two. (d) The low energy excitation feature in single chain $SrCuO_2$, measured from $Q = 2\pi$ to 3π, exhibits a similar dispersion to the ladder peaks but has one dispersive peak.

(ladder "rung"). The low energy features have no measured dispersion along this direction, but appear to come slightly closer together near $Q = \pi/2$ and $Q = \pi$.

An image plot with momentum transfer across the full second ladder Brillouin zone is shown in Fig-4(c). The low energy features of SCO are roughly symmetric about $Q = 2\pi/c_L$, and no signal corresponding to the chain periodicity is observed. Dispersion over the second half of the second Brillouin zone ($Q = 2\pi$ to 3π) appears to be smaller than from $Q = \pi$ to 2π by less than 0.1 eV, which is of the same order as previously estimated inter-ladder coupling ($\sim 0.026eV$[15], yellow lines in Fig-1(a)), and may be due to the true $4\pi/c_L$ ladder plane periodicity. Dispersion in quasi-two dimensional compounds, such as Nd_2CuO_4 shown in Fig-4(d), is much smaller in the Cu-O bond direction, and is accompanied by a large damping of intensity across the Brillouin zone. By contrast, the corner-sharing single chain compound is thought to have only a single $\sim$1.1eV dispersion low energy charge excitation peak, with enhanced spectral weight at the Brillouin zone edge[11]. The compound Nd_2CuO_4 is chosen to represent the 2D cuprates because its low energy signal has never been measured before, and it is more significant with respect to the dimensionality crossover because it does not have an apical oxygen, unlike $La_{2-x}Sr_xCuO_4$.

Dispersion and peak intensity of the two features observed in SCO fall between those of 1D and 2D systems, implying a continuous transition between distinct 1D and 2D charge dynamics. In order to understand the results, we consider a variation of the 1D strong coupling limit ($U \gg t$) in which the excited electron hole pairs of a single band Hubbard model are unaffected by local spin [14]. In a renormalized picture, the dispersion of free "hole" and "double occupancy" quasiparticles that are separated from the spin background is given by $2t_L cos(kc_L) \pm t_\perp$ in the ladder and $2t_{ch} cos(kc_{ch})$ in the chain, with t_L representing nearest neighbor hopping in the ladder structure $\hat{c}$ axis direction and $t_\perp$ giving hopping across the ladder rungs. These bands are expected to be split by the onsite Coulomb interaction (Hubbard U). Such a band structure has been used to interpret the infrared spectrum, which probes charge excitations with $Q = 0$ [15]. We have plotted renormalized band structure and the allowed regions of interband transition spectral weight for such a model in Fig-5(a-b), with the dispersion of low energy RIXS features overlaid for comparison. Dispersion of the two lowest energy RIXS features follows the lower edges of the first and third continua. It has been analytically shown that in a 1D strong coupling model in the presence of intersite Coulomb interaction, *most* spectral weight concentrates near the lower edge of the continuum [14]. Since the ladder can be modeled as weakly interacting chains, spectral weight pile-up near the lower edges of the continuum consistent with the observed behavior can be interpreted as a consequence of strong Coulomb interaction.

Now we discuss the apparent absence of the chain layer band. The absence or weakness of the signal can be understood in the strong coupling limit too. The charge transfer energies in the ladder and chain substructures of $Sr_{14}Cu_{24}O_{41}$ are similar in magnitude (on the order of 1.5-3 eV [6, 15]), so the spectral intensities from ladder and the chain are expected to overlap in the energy window of interest. We mark the regime of the lower-energy signal from the chain in Fig-5 based on Ref[6, 15]. In the strong-coupling limit, the *spectral intensity* of charge response is proportional to the square of the scaled hopping parameters ($N_q \sim t^2/U^2$) [14]. The chain substructures are edge-shared (Fig-1), therefore direct hopping is very small compared to hopping along the "legs" of the ladder, leading to a relatively large contribution of spectral intensity due to the Cu_2O_3 ladder substructure over

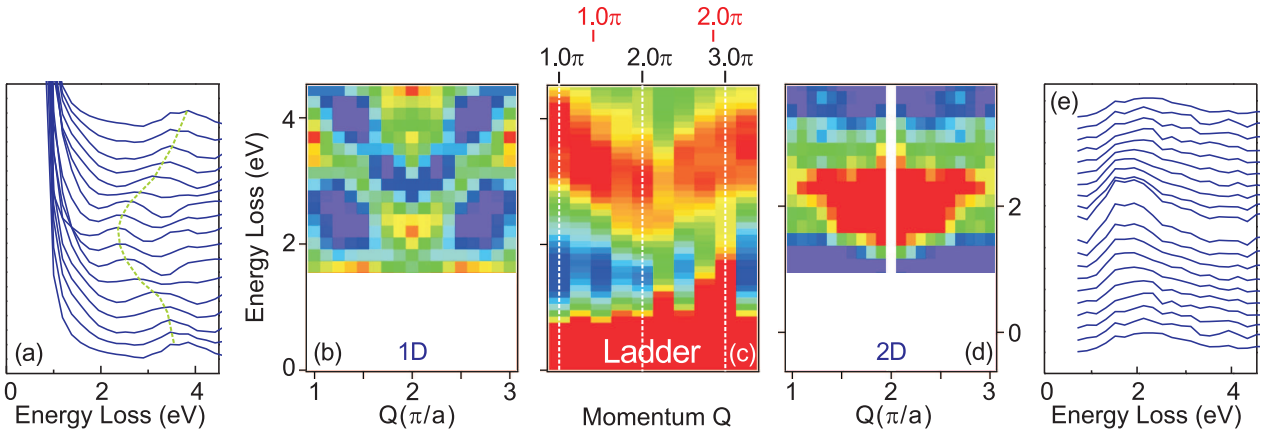

FIG. 4: **Q-dependence and lattice dimensionality**: Inelastic charge response (c-E_1 RIXS) spectral weight across the Brillouin zone for (a-b) quasi-1D $SrCuO_2$, (c) $Sr_{14}Cu_{24}O_{41}$ and (d-e) quasi-2D Nd_2CuO_4. Spectra were measured from $Q = 2\pi$ to 3π (a-b), $Q = \pi$ to 3.25π (c), and π to 2π (d-e), and images in (b) and (d) are symmetrized about $Q = 2\pi$. In panel (c), red ticks mark the positions in the chain BZ and black ticks mark positions in the ladder BZ. Low energy spectral weight is found to be periodic with the ladder.

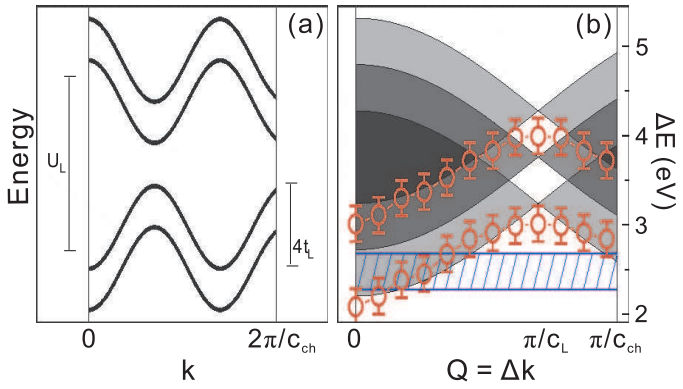

FIG. 5: (a) Renormalized band structure of the ladder. Splitting is due to rung coupling. (b) Energy range of all possible two particle excitations (grey) in the ladder based on panel (a). Experimental peak positions determined from Fig-(3) are overlaid (red circles) for comparison. Hatched regions (2.3 to 2.7 eV) mark the experimental boundaries for the two particle spectrum of the chain layer expected to overlap with the ladder continuum based on Ref[15].

the CuO_2 chain-containing planes. This is further confirmed by the large magnitude ($\sim$ 1eV) of the measured dispersion (Fig-3). The weakness of hopping integral for the edge-sharing chain suggests that the associated mode would be very weakly dispersive (essentially flat) in contrast to the observed Q-dependence of the experimental peaks in the relevant charge-transfer energy range. Finally, the experimentally observed periodicity of dispersion relations correspond to $2\pi/c_L$ but not $2\pi/c_{ch}$ (Fig-4(c)). Therefore, the predominant contribution is due to the ladder layer.

The experimentally observed charge dynamics along the "rung" direction can be understood in the following qualitative scheme: Since the inter-ladder virtual electron hop is via a ninety degree exchange path its magnitude is small compared to the intra-ladder hopping, so the pair excitation near the gap edge is mainly confined to the individual ladder units and propagates (hence dispersive) along the leg direction of the ladder units. This is consistent with our finding of the lack of dispersion of excitation modes along the $\hat{a}$-direction (Fig-3(c)) as well as weaker spectral intensity. This scenario and the fact that dispersion behavior is similar to the corner-sharing chain compound (Fig-3(d)), at least near the insulating gap edge, support the quasi-one (river-like) dimensionality of the collective charge excitation modes in $Sr_{14}Cu_{24}O_{41}$. Such collective charge dynamics are significantly different from what is typically observed in most two dimensional cuprates such as the Nd-Ce-Cu-O or La-Sr-Cu-O systems [12] where the dispersion anisotropy is much weaker (measured under the same polarization and scattering geometry condition at the analogous incident energy). Charge dynamics in ladders also differ in details from that in 1D, in the sense that no half-periodic spinon-like on-set [11] is observed. This is possibly due to suppression of charge-spin separation in the ladder geometry from dimer-coupling in the rungs [6].

In summary, our results demonstrate that the low-energy excitation modes at the gap edge in $Sr_{14}Cu_{24}O_{41}$ predominantly originate from the ladder containing quasi-two dimensional layers. The measured dispersion relations of the low-energy modes and distribution of spectral weight fall between what is typically observed in similar 1D and 2D systems, suggesting distinct physics associated with intermediate dimensionality. We suggest that traits of such an anisotropic mode spectrum can be described in the strong-interaction limit with lateral con-

finement of electron-hole pairs although a full quantitative description requires multi-band model Resonant-IXS calculation with a very large unit cell [16]. We speculate that charge dynamics in the doped system will deviate greatly from this behavior under high pressures where cuprate-like (2D) superconductivity is achieved. Perhaps the application of pressure makes it more two dimensional enhancing the pairing correlations.

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